

DRAWINGS ATTACHED

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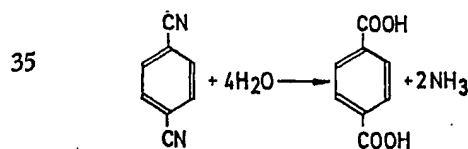
(54) PRODUCTION OF AROMATIC CARBOXYLIC ACIDS

(71) We, THE LUMMUS COMPANY, 1515 Broad Street, Bloomfield, New Jersey 07003, United States of America, a corporation organised and existing under the laws of the State of New Jersey, United States of America, do hereby declare the invention, for which we pray that a patent may be granted to us, and the method by which it is to be performed, to be particularly described in and by the following statement:—

The invention relates to the production of an aromatic mono- or poly-carboxylic acid by the hydrolysis of an aromatic mono- or poly-nitrile.

The catalytic conversion of aromatic hydrocarbons to aromatic polynitriles, as xylenes to benzodinitriles, is known. Suitable conversions are described in British Specification No. 1,138,696. Hydrolysis of the benzodinitriles to the corresponding benzene dicarboxylic acids is described in British Specification No. 1,162,359. By integrating the catalytic conversion and the hydrolysis process, it is possible to prepare a polynitrile, purify the polynitrile and then hydrolyse the purified polynitrile. For example, p-xylene may be converted to terephthalonitrile which is then purified and the purified nitrile is subjected to hydrolysis. Terephthalic acid is recovered from the resulting hydrolysis product for use in the manufacture of resins and fibres.

The overall reaction involved in the hydrolysis of terephthalonitrile is:



Processes at present in use for hydrolysing nitriles to the corresponding acids are carried out in two steps. First, the nitrile is partially hydrolysed to a salt, ester, or amide, which is then again hydrolysed to the

carboxylic acid. Such processes can be costly to use because they require two sets of hydrolysing equipment, including vessels, pipes and instruments.

It is an object of the invention to provide a single step process for the production of an aromatic mono- or poly-carboxylic acid by the hydrolysis of its corresponding aromatic mono- or poly-nitrile.

According to the invention there is provided a process for the production of an aromatic carboxylic acid by the hydrolysis of an aromatic nitrile which comprises (a) maintaining an aqueous slurry of the aromatic nitrile in contact with a catalyst which is an alkali metal or alkaline earth metal hydroxide, carbonate or salt of an aromatic carboxylic acid or ammonium salt of an aromatic carboxylic acid at a temperature of from 300° F to 600° F for a period sufficient to hydrolyse a substantial portion, preferably substantially all of the nitrile, (b) stripping ammonia from the hydrolysate product by countercurrently contacting the hydrolysate with steam, and (c) cooling the stripped hydrolysate to a temperature at which the aromatic carboxylic acid separates out. The stripped hydrolysate is preferably cooled to a temperature of from 100° F to 300° F. When the catalyst is a salt of an aromatic carboxylic acid, it is preferably a salt of the acid corresponding to the nitrile.

The process according to the invention produces carboxylic acids suitable for use in many applications. Where acids of extremely high purity are required, for example, as monomers for producing polymeric fibres in which discolouration resulting from the presence of minor amounts of nitrogen-containing impurities is undesirable, the acid may be further purified by repulping the acid with water and a minor amount of said catalyst, holding the mixture so formed at a temperature higher than the temperature of the first hydrolysis step (a) but less than 650° F for a period sufficient to hydrolyse the hydrolysable components thereof, and then cooling the hydrolysis product to a temperature

[Price 25p]

of from 100° F to 300° F so as to separate out the aromatic carboxylic acid which may then be recovered.

5 In this second hydrolysis step, the undesirable nitrogen-containing compounds, which are mainly partially hydrolysed forms of the nitrile, are further hydrolysed to the carboxylic acid.

10 In the first hydrolysis, step (a), steam at a temperature of from 300° F to 600° F may be added to the heated slurry and steam at a temperature of from 350° F to 650° F may be added to the repulped acid in the second hydrolysis step.

15 The process according to the invention permits production of carboxylic acids from nitriles using less equipment than has hitherto been required. Consequently, the utility requirements are also lowered. Additionally, 20 fewer steps are required for purification, and the purity of the product is more easily controlled.

Methods of carrying out the invention are illustrated by way of example in the accompanying drawings in which

25 Figures 1 and 2 are simplified flow diagrams for the hydrolysis of terephthalonitrile to terephthalic acid and for recovery of the latter.

Referring now to Figure 1, which depicts 30 a preferred embodiment of the invention, as used for producing terephthalic acid of high purity, terephthalonitrile which is substantially free from impurities, particularly hydrocarbons, aromatic and aliphatic mononitriles and dinitriles, ketones, alcohols and aliphatic acids, is passed from line 11 into a slurry 35 tank 10 equipped with an agitator 12. Water containing a minor amount of catalyst, such as dipotassium terephthalate, is passed from a process source, described hereinafter, through 40 line 13 into the slurry tank 10. Fresh or make-up catalyst solution can also be added to line 13 from line 14. A slurry of terephthalonitrile, water and catalyst is formed in 45 the tank 10 and is heated therein to approximately 200° F. Steam from line 15 is passed into tank 10 and serves to provide heat for the slurry.

50 The heated slurry is removed from tank 10 through a line 16, reciprocating pump 17 and line 18 wherein it is combined with steam at approximately 430 psia in line 19 to raise the temperature of the slurry to the desired hydrolysis temperature, such as about 55 395° F, the slurry being then passed into a primary hydrolyser 20. The temperature in hydrolyser 20 is from 300° F to 600° F and is preferably in the range 350—540° F.

60 Pressure in hydrolyser 20 is determined by the temperature of the liquid at the top thereof and by the partial pressure of the ammonia which is evolved during the hydrolysis. Typical pressures range from 100 to 2000 pounds per square inch absolute (psia). 65 In the primary hydrolyser 20, terephthalonitrile is converted to terephthalic acid, the

degree of conversion being dependent upon the temperature therein, the residence time and the catalyst used. Such operating variables are discussed in detail hereinafter. In this 70 illustration the potassium cation concentration in the charge to hydrolyser 20 is about 5 mols per 100 mols of nitrile group of the terephthalonitrile charge, and the residence time is about 6 hours. 75

In a continuous process, the catalytic cation combines with the acids formed in hydrolyser 20 to form the corresponding salts. Thus, with potassium carbonate, potassium hydroxide or sodium carbonate used initially as a catalyst, the potassium or sodium forms salts 80 with acids formed in the process and is recycled in the form of the potassium or sodium salts of terephthalic acid and of terephthalamic acid. It is advantageous to hydrolyse only a 85 substantial portion of the terephthalonitrile to terephthalic acid; in such case, a lower conversion is realised, but available for recycle as catalysts are the potassium salts of terephthalic acid and of terephthalamic acid and 90 the ammonium salts of the same acids together with terephthalamide. Ammonium salts of terephthalic acid and terephthalamic acid also act as catalysts in the hydrolysis of aromatic mono- and polynitriles to their corresponding acids. 95

Ammonia is stripped from the hydrolysis reaction products by steam added through line 21 and passed countercurrent to the reaction 100 products, and is removed from hydrolyser 20 through overhead line 22. The ammonia can be charged, for example, to a reactor (not shown) with p-xylene for the production of terephthalonitrile.

Terephthalic acid, formed by hydrolysis in 105 hydrolyser 20, is removed as a slurry together with other hydrolysis products through line 23, controlled by valve 24 therein, to a flash vessel 25. Steam is removed from vessel 25 through line 15. As steam is flashed from 110 the hydrolysis product, the temperature thereof is lowered, for example to 240° F.

The cooled hydrolysis products are passed 115 from the vessel 25 through a line 26 to a centrifuge 27. In the centrifuge, terephthalic acid is separated from materials in solution in the hydrolysis products.

If the product is suitably pure for its intended use, the crystals are recovered from the centrifuge and purified by conventional 120 means (not shown). If, however, a product of extremely high purity is required, additional purification is accomplished as follows.

The terephthalic acid, now containing as 125 impurities only some partially hydrolysed products of terephthalonitrile, is passed from the centrifuge 27 through line 28 to a second slurry tank 29 which is equipped with stirrer 30. The terephthalic acid is repulped in tank 29 with water and a minor amount of cata- 130

lyst from line 31. Steam is passed from line 32 into a lower portion of the tank 29 to heat the slurry which forms therein. The temperature maintained in tank 29 is from 150° F to 220° F. The resulting heated slurry in tank 29 is passed through line 33, reciprocating pump 34 and line 35 to a heat exchanger 36 wherein it extracts heat, and thence through line 37 to a secondary hydrolyser 38. Steam under high pressure, for example 885 psia, in line 39 is passed into line 37 to join with and to heat further the material in line 37 before it is introduced in the hydrolyser 38.

In the hydrolyser 38, the temperature is maintained above that which is employed in the primary hydrolyser 20 but not more than 650° F, the temperature in the hydrolyser 38 being in any case preferably above 350° F. The pressure in the hydrolyser 38 is essentially the vapour pressure of water at the temperature of the contents of hydrolyser 38. Any ammonia liberated during hydrolysis is removed from hydrolyser 38 together with steam through vent 40. Other hydrolysis products, including terephthalic acid, are removed from the hydrolyser 38 through bottom line 41 and heat exchanger 36, and pass through line 42 controlled by valve 43 to a flash vessel 44. In the latter, steam is flashed via line 32 to the slurry tank 29. As steam is flashed from the vessel 44, the temperature of the hydrolysis products therein falls, for example to 240° F. The cooled hydrolysis products are essentially terephthalic acid crystals and traces of water-soluble ammonium and catalytic cation salts of terephthalic acid and terephthalamic acid, and are passed from the vessel 44 through line 45 to a second centrifuge 46.

In centrifuge 46, terephthalic acid is separated from other hydrolysis products. Water is passed through line 47 into the centrifuge 46 to wash the terephthalic acid to remove any adherent traces of soluble impurities therefrom. Substantially pure terephthalic acid crystals are passed from the centrifuge 46 through line 48 to a dryer 49 for the removal of water therefrom, and the dried crystals are then removed through line 50.

Aqueous solution separated in centrifuge 46 is passed through line 51 to a receiving vessel 52 from which the solution is pumped, by pump 53 in line 54, in part to line 31 and in part to line 55. That portion diverted to line 31 is used in the repulping of terephthalic acid in tank 29. The remaining portion is passed through line 55 to the centrifuge 27 wherein it serves as a wash medium for the terephthalic acid cake therein.

The aqueous materials separated in centrifuge 27 are passed through line 56 to a receiver 57. They are removed from the latter through discharge line 58, from which a portion can be removed through purge line 59. The remainder in line 58 is pumped via pump 60 in line 13 to the slurry tank 10.

A typical example in which the process sequence is as shown in Figure 1, is given in the following Example 1. All quantities are expressed in moles per hour, unless otherwise specified. Terephthalonitrile is abbreviated to "TPN" and terephthalic acid to "TPA" for convenience.

EXAMPLE 1

Fresh charge to Slurry Tank 10
TPN, line 11 75.3 75

Feed to Primary Hydrolyser 20
Water, line 13 1869.6
TPA, line 13 0.1
Diammonium terephthalate, line 13 20.3
Monoammonium terephthalamate, line 13 2.3 80
Dipotassium terephthalate, line 13 3.0
Monopotassium terephthalamate, line 13 6.8
TPN 75.3 85

Steam to Hydrolyser 20
Charge, line 18 414.1
Stripping, line 21 95.9

Hydrolyser (20) Overhead in line 22
Ammonia 150.6 90
Water 361.7
Water reacted in hydrolyser (20) 301.2

Hydrolyser (20) Product in line 23
TPA 75.4
Diammonium terephthalate 20.3 95
Monoammonium terephthalamate 2.3
Dipotassium terephthalate 3.0
Monopotassium terephthalamate 6.8
Water 1302.6

Centrifuge (27) Product in Line 28 100
TPA 75.3
Water 462.9

Centrifuged Solution in Line 56
TPA 0.1
Diammonium terephthalate 20.3 105
Monoammonium terephthalamate 2.3
Dipotassium terephthalate 3.0
Monopotassium terephthalamate 6.8
Water 1188.4

Primary Flash Vapour in Line 15 267.2 110

Centrifuge (27) Cake Wash in Line 55
TPA 0.02
Water 615.9

Repulp Slurry in Tank 29
TPA 75.4 115
Water 1233.3

Steam to Secondary Hydrolyser (38) in Line 39 125.7

Secondary Hydrolyser (38) Product in Line 41		
TPA	75.4	
Water	1359.0	
Secondary Flash Vapour in Line 32 69.4		
5 Centrifuge (46) Product in Line 48		
TPA	75.3	
Water	462.9	
Centrifuge (46) Cake Wash in Line 47		
Water	490.3	
10 Centrifuged Solution in Line 51		
TPA	0.07	
Water	1317.5	

Thus, there is essentially mole per mole conversion of terephthalonitrile to terephthalic acid, with the latter being of a high degree of purity.

Figure 2 illustrates a modified process operation, in which higher hydrolysis temperatures are employed. Since many features of Figure 2 are the same as those of Figure 1, only the modifications are described in detail in the interests of brevity. The cation used in this example was sodium rather than potassium. Slurry tank 100 received terephthalonitrile from line 101. Tank 100 is equipped with a stirrer 102 and with a steam coil 103 which provides heat to the contents of the tank. Water and catalyst recycled in the process are delivered to tank 100 from line 104. Make-up water and catalyst can be added to line 104 via line 105.

Heated slurry is passed from tank 100 through line 106, reciprocating pump 107 and line 108 in which is located a heat exchanger 109. In passing through heat exchanger 109, the slurry is heated from 260° F to 460° F. High pressure saturated steam, 885 psia and 530° F in line 99 is added to line 108, such that the slurry and steam therein are passed together into primary hydrolyser 110. Hydrolysis of the terephthalonitrile occurs in hydrolyser 110, at substantially 500° F, 800 psia and a residence time of about 6 hours. Ammonia is stripped from the hydrolysis products by additional steam taken from line 99 through line 111, and contacted countercurrently with the hydrolysis products, the stripped ammonia being removed from hydrolyser 110 through line 112.

The balance of the hydrolysis products are passed from hydrolyser 110 through discharge line 113 to the heat exchanger 109 wherein heat is extracted from the products. The hydrolysis products are then passed through line 114, controlled by valve 115, to a flash vessel 116. Steam is removed in vessel 116 through line 117, the temperature of the remaining hydrolysis products being lowered thereby to substantially 210—220° F. The

cooler hydrolysis products are passed through line 118 to centrifuge 119, wherein terephthalic acid is separated from other products.

Terephthalic acid is taken from centrifuge 119 through line 120 to a second slurry tank 121 which is equipped with an agitator 122. The terephthalic acid is repulped in tank 121 with water and a minor amount of catalyst from line 123. The repulped acid in tank 121 is at a temperature of substantially 200° F, when it is taken through line 124, reciprocating pump 125 and line 126 to heat exchanger 127 wherein it is heated to substantially 420° F. The heated material is then passed through line 128 in which it receives high pressure, saturated steam at substantially 530° F and 885 psia from line 129 before passing into a secondary hydrolyser 130.

Hydrolysis conditions in hydrolyser 130 include a temperature of substantially 520° F, a pressure of substantially 810 psia and a residence time of substantially one hour. Any ammonia formed is removed through line 131. Other hydrolysis products are passed from hydrolyser 130 through line 132 to heat exchanger 127 wherein heat is extracted. Cooled hydrolysis products are then passed through line 133 and equipment 134—145 which duplicates equipment 43—54 of Figure 1.

Aqueous solution separated in centrifuge 137 and passed to line 145 is passed, in part, through line 123 to tank 121, and in part, through line 104 to tank 100.

Aqueous materials separated in centrifuge 119 are passed through line 146 to tank 147, wherein they are combined with steam from line 117, flashed from vessel 116, and with steam from line 148. The aqueous mixture in tank 147 is removed through line 149; a portion thereof can be purged through line 150 and the balance can be pumped by pump 151 in line 152 to line 104.

The operations shown in Figure 2 are illustrated by the following typical example.

EXAMPLE 2

Fresh Charge to Slurry Tank 100		
TPN, line 101	75.3	
Feed to Primary Hydrolyser 110		
Water, line 104	5144.9	110
TPA, line 104	0.1	
Monosodium terephthalamide, line 104	3.8	
Disodium terephthalate, line 104	1.9	115
Terephthalamide, line 104	0.8	
TPN	75.3	
Steam to Hydrolyser 110		
Charge line 108	298.4	
Stripping, line 111	79.4	120

	Hydrolyser (110) Overhead in Line 112	
	Ammonia	150.6
	Water	853.4
	Water Reacted in Hydrolyser 110	301.2
5	Hydrolyser (110) Product in Line 113	
	TPA	75.4
	Disodium terephthalate	1.9
	Monosodium terephthalamate	3.8
	Terephthalamide	0.8
10	Water	3990.3
	Centrifuge (119) Product in Line 120	
	TPA	75.3
	Water	462.9
	Centrifuged Solution in Line 146	
15	TPA	0.1
	Disodium terephthalate	1.9
	Monosodium terephthalamate	3.8
	Terephthalamide	0.8
	Water	3527.4
20	Repulp Slurry in Tank 121	
	TPA	75.4
	Water	2777.8
	Steam to Secondary Hydrolyser (130) in Line 129	411.1
25	Secondary Hydrolyser (130) Product in Line 132	
	TPA	75.4
	Water	3188.9
30	Secondary Flash Vapour in Line 148	313.0
	Slurry Charge to Centrifuge 137	
	TPA	75.4
	Water	2875.8
35	Centrifuge (137) Cake Wash in Line 138	
	Water	829
	Centrifuge (137) Product in Line 139	
	TPA	75.3
	Water	462.9
40	Centrifuged Solution in Line 123	
	TPA	0.04
	Water	2314.8
	Centrifuged Solution in Line 104	
	TPA	0.02
	Water	926.7
45	Here again, there is essentially mole per-mole conversion of terephthalonitrile to terephthalic acid.	
50	While the process illustrated in Figure 2 is effective for the purpose of efficiently obtaining terephthalic acid of a high degree of purity, the process illustrated in Figure 1 is	

preferred. Thus, the hydrolysis of terephthalonitrile can be conducted at lower temperatures, lower pressures and with higher terephthalonitrile concentrations than shown with Figure 2. By operating at such temperatures, pressures, and concentrations, the initial hydrolyser (e.g. 20) can be smaller in size and less expensive than that required in connection with Figure 2.

The invention has been specifically illustrated by the conversion of terephthalonitrile to terephthalic acid. It is to be understood, however, that the invention can also be employed for the hydrolysis of other aromatic mono- and polynitriles to their corresponding aromatic mono- and poly-carboxylic acids. For example, the following hydrolyses can be effected: benzonitrile to benzoic acid; toluenitrile to toluic acid; o-phthalonitrile to phthalic acid; isophthalonitrile to isophthalic acid; p-cyanobenzamide to terephthalic acid, and 1,2,4 - benzenetrinitrile to trimellitic acid.

The nitrile charged in the process can be pure or can be contaminated with other materials which are formed in the preparation of the nitrile. However, it will be recognised that more efficient operation is realised with a nitrile charge substantially free of contaminants.

As indicated, catalysts employed in the hydrolysis are alkali or alkaline earth metal hydroxides, carbonates and salts of the aromatic mono- or poly-carboxylic acids formed in the hydrolysis and ammonium salts of said acids. In the primary hydrolysis, the catalytic cation concentration is suitably from 1 to 30 moles, preferably 2—10 moles, per 100 moles of functional nitrile group in the charge. In the secondary hydrolysis, the catalytic cation concentration may vary from 0.1 to 30 moles per mole of hydrolysable functional group.

Temperatures maintained in the primary hydrolysis vary from 300° F to 600° F, the preferred temperatures being in the range 350—540° F. In the secondary hydrolysis, a higher temperature is employed with a maximum of 650° F, the secondary hydrolysis being preferably carried out at a temperature in the range 450—600° F. Generally, a temperature differential of substantially 130° F is maintained.

Pressures in the primary and secondary hydrolysers may range from 100 to 2000 psia.

Residence time for the primary hydrolysis is suitably from 1 to 10 hours, preferably about 6 hours. In the secondary hydrolysis, a shorter residence time is required, for example 0.01 to 1 hour.

It is desirable to hydrolyse only a substantial portion of the nitrile to the corresponding acid in the primary hydrolyser whether or not the secondary hydrolyser is to be employed. Since the nitrogen-containing impuri-

ties, such as amides and terephthalamic acid, are soluble in water whereas terephthalic acid is not, the acid recovered after centrifuging and washing will be substantially pure even if the secondary hydrolyser is not utilised, and will be suitable for most uses, except when an extremely pure acid is required. The wash water, containing the nitrogen-containing impurities and salts of the acid, is preferably recycled back to the primary hydrolyser.

WHAT WE CLAIM IS:—

1. A process for the production of an aromatic carboxylic acid by the hydrolysis of an aromatic nitrile, which comprises (a) maintaining an aqueous slurry of the nitrile in contact with a catalyst which is an alkali metal or alkaline earth metal hydroxide, carbonate or salt of an aromatic carboxylic acid or ammonium salt of an aromatic carboxylic acid at a temperature in the range 300°—600° F for a period sufficient to hydrolyse a substantial portion of the nitrile, (b) stripping ammonia from the hydrolysate, by countercurrently contacting the hydrolysate with steam, and (c) cooling the stripped hydrolysate to a temperature at which the aromatic carboxylic acid separates out.
2. A process according to claim 1, in which the salt is a salt of the acid corresponding to the nitrile.
3. A process according to claim 1 or claim 2, in which substantially all of the aromatic nitrile is hydrolysed.
4. A process according to any one of the preceding claims, in which the stripped hydrolysate is cooled in step (c) to a temperature of from 100° F to 300° F so as to cause the aromatic carboxylic acid to separate out.
5. A process according to any one of the preceding claims, which includes (d) repulping the aromatic carboxylic acid from step (c) with water and a minor amount of said catalyst; (e) holding the mixture formed in step (d) at a temperature higher than the temperature used in step (a) but not higher than 650° F for a period sufficient to hydrolyse the hydrolysable components thereof; and (f) cooling the hydrolysis product of step (e) to a temperature of from 100° F to 300° F so as to separate out the aromatic carboxylic acid.
6. A process according to claim 5, in which the catalyst cation concentration in step (d) is from 0.1 to 30 moles per mole of hydrolysable functional group.
7. A process according to claim 5 or claim 6, in which the repulped carboxylic acid is heated in step (d) to a temperature of from 350° F to 650° F, and steam at a temperature of from 350° F to 650° F is added thereto.
8. A process according to any one of the preceding claims, in which the aqueous slurry in step (a) is heated to a temperature of from 300° F to 600° F, and steam at a temperature of from 300° F to 600° F is added to the heated slurry.
9. A process according to any one of the preceding claims, in which the hydrolysis of the nitrile is carried out at a temperature in the range 350°—540° F.
10. A process according to any one of the preceding claims, in which the nitrile is a mononitrile and the carboxylic acid is a monocarboxylic acid.
11. A process according to any one of claims 1 to 9, in which the nitrile is a polynitrile and the carboxylic acid is polycarboxylic acid.
12. A process according to any more of claims 1 to 9, in which the nitrile is a dinitrile and the carboxylic acid is a dicarboxylic acid.
13. A process according to any one of claims 1 to 9, in which the nitrile is a trinitrile and the carboxylic acid is a tricarboxylic acid.
14. A process according to any one of claims 1 to 9, in which the nitrile is terephthalonitrile.
15. A process according to claim 14, in which the catalyst is an alkali metal salt of terephthalic acid.
16. A process according to claim 14, in which the catalyst is ammonium terephthalate.
17. A process according to claim 14, in which the catalyst is ammonium terephthalate.
18. A process according to any one of claims 1 to 13, in which the catalyst is an alkali metal or alkaline earth metal salt of the aromatic carboxylic acid.
19. A process according to any one of claims 1 to 14, in which the catalyst is sodium carbonate or potassium carbonate.
20. A process according to any one of claims 1 to 14, in which the catalyst is potassium hydroxide.
21. A process according to any one of claims 5 to 20, in which the catalyst cation concentration in the primary hydrolysis of the nitrile is from 1 to 30 moles per 100 moles of functional nitrile group.
22. A process according to claim 21, in which said concentration is from 2 to 10 moles per 100 moles of functional nitrile group.
23. A process according to any one of claims 1 to 5, substantially as hereinbefore described with reference to Figure 1 or Figure 2 of the accompanying drawings.
24. A process for the production of an aromatic carboxylic acid from the corresponding nitrile, substantially as hereinbefore described

with reference to Example 1 or Example 2.

25. An aromatic carboxylic acid whenever obtained by the process claimed in any one of the preceding claims.

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FIGURE 1

